

TITLE OF INVENTION

Tantalum Water Target Body for Production of Radioisotopes

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] Not Applicable

5 STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR
DEVELOPMENT

[0002] Not Applicable

BACKGROUND OF THE INVENTION

1. Field of Invention

10 **[0003]** This invention relates to the field of target assemblies for use with
accelerators for the production of radioisotopes. More particularly, this invention
pertains to target assemblies, which have less than ideal thermal conductivity,
having internal cooling channels and thermally optimized target chambers.

2. Description of the Related Art

15 **[0004]** Positron Emission Tomography (PET) is a powerful tool for diagnosing
and treatment planning of many diseases wherein radioisotopes are injected into a
patient to diagnose and assess the disease. Accelerators are used to produce the
radioisotopes used in PET. Generally, an accelerator produces radioisotopes by
accelerating a particle beam and bombarding a target material, housed in a target
20 system, with the particle beam.

[0005] Several factors must be considered when developing a target system
for the production of radioisotopes. In the case of gas or liquid targets, the target
material must be maintained at an elevated pressure during bombardment to
compensate for the effects of density reduction of the target material due to
25 heating/expansion/phase change (boiling). Further, it is desirable to operate at

higher beam currents to increase production of the radioisotopes. Because of the amount of heat generated during bombardment, cooling the target material and other components of the target system is of significant importance.

[0006] Enriched water targets are used for the commercial production of the short lived ($t_{1/2} = 109.8$ minutes) positron emitter fluorine-18 (^{18}F) for use as a tracer for Positron Emission Tomography (PET). The desired isotope is produced by proton bombardment of ^{18}O enriched water (enrichment typically above 95%), using the $^{18}\text{O}(\text{p},\text{n})^{18}\text{F}$ reaction. The ^{18}F isotope is used to produce fluorodeoxyglucose (FDG), which, when introduced within a patient, is used to map metabolic rates in the patient.

[0007] The cost of the enriched water and the short half-life of ^{18}F drive competing constraints on the target design. In order to overcome decay losses the target production must be maximized. This requires the target assemblies be designed for maximum operating current, which also increases ionization heating of the bombarded water. In order to minimize cost of reagents (specifically the expensive enriched water), the target assemblies necessarily have a small volume (<2 ml). Typical volume averaged power density in such targets is 400 W/cc. However peak power densities can be as much as two orders of magnitude greater.

[0008] Figures 1 and 2 illustrate perspective views of a prior art target assembly **110** showing the front surface **112** and rear surface **212**, respectively. Figure 3 is a cross-sectional view of the target assembly **110**. The target assembly **110** has a front face **112**, which is adapted to connect to an accelerator or cyclotron. The target assembly **110** has a cylindrical body which fits into a cylindrical slot which supplies cooling water to the target assembly **110**. The target assembly **110** also has a rear face **212**, which has connections **220**, **222** for the enriched water and openings for securing **214**, **216** the target assembly **110**.

[0009] The prior art target assembly **110** includes a target chamber **104** encased in silver and having cooling channels **102, 104, 202, 204, 302, 304** along the outside surface of the target assembly **110**. Typically, cooling water flows into the channel **104** on the bottom of the target assembly **110**, through the channels
5 **302, 304** along the circumference of the target assembly **110** and the channels **202, 204** along the rear surface **212** of the target assembly **110**, and collecting in the channel **102** on the top of the target assembly **110**, where it is removed and run through a heat exchanger to remove the collected heat.

[0010] The prior art target assembly **110** includes a target chamber **104**,
10 which is filled with enriched water via an inlet port **220** on the back side **212**. The target chamber **104** is sealed with a window **310** adjacent the front face **112**. The inlet port **220** feeds an inlet channel **106**, through which the enriched water enters and fills the target chamber **104**. The air pushed out of the target chamber **104** exhausts through the outlet port **222**. Before being irradiated, the enriched water
15 completely fills the target chamber **104**.

[0011] The prior art target assembly **110** is fabricated from a silver ingot and operates at approximately 600 watts (10 MeV protons at 60 μ A) on the target water. Irradiation of ^{18}O -water in silver target bodies with proton beam currents higher than 30 μ A generally leads to formation of gray or black colloids which
20 frequently clogs the ^{18}F ion delivery lines. More importantly, the reactivity of the ^{18}F ion thus obtained is severely diminished. A model of the prior art target assembly **110** has been generated. This model of the external coolant cycle exposed inefficient cooling mechanisms, opportunities for coolant dryout, and likelihood of flow instabilities.

[0012] Silver target assemblies **110** oxidize under the conditions seen in a high pressure water target, and eventually this oxidation leads to failure of the system, both through increased temperature drops through the oxide, sequestering of the fluoride product on the oxide surface, and oxide particles fouling the product capillary tubing and subsequent synthesis into the desired tracer. At high
25 currents, such as 40-60 μ A, the silver target holders are typically only usable for
30

20 to 30 runs to create radioisotopes such as Flourine-18 before being too contaminated for further use to maintain sufficiently pure radiochemicals. At that point the target assembly must be removed from the accelerator and cleaned to recover functionality.

5 **[0013]** Various factors effect the production of radioisotopes from liquid targets with low energy accelerators. One such factor includes the configuration of the holding assemblies that retain the liquid target during the irradiation process. The holding assemblies must withstand severe environments created during the irradiation process and also enable the production of contaminant-free
10 radiochemicals. When the liquid target is irradiated, the proton beam quickly heats the liquid target and creates high pressure within the target holder. The target holder must be capable of withstanding the elevated pressures without rupturing and without removing too much energy from the proton beam. Conventional liquid target holders have a thin front window through which the
15 proton beams must pass before hitting the liquid target. Thicker windows are desirable to withstand the pressures generated from heating the liquid, but the thicker windows provide more mass through which the proton beam must pass before reaching the target. Accordingly, the thicker windows absorb more beam energy, thereby decreasing the effectiveness of the proton beam. When a low energy
20 beam is used, it is highly desirable to ensure that as much energy remains in the proton beam as possible by the time it reaches its liquid target to maximize the beam's efficiency for irradiating the liquid target. So, while the strength of the thick window is desired, the resulting energy decrease in the beam is not.

25 **[0014]** Another factor includes providing a liquid target that will fully absorb the remaining energy of the proton beam. As the proton beam is passed into the target holder and the target liquid, the target liquid must have a sufficient depth or thickness so as to fully absorb the particles from the beam. If the proton beam passed completely through the liquid target and the target holder, the particle beam could create a radioactive environment external to the holding assembly.

[0015] Another significant factor in forming the radioisotopes or radiochemicals is controlling the target liquid's temperature during the irradiation process. When the proton beam bombards the target liquid, the temperature of the target liquid quickly increases. Heat must be efficiently drawn from the target liquid to maximize the effective density of the target liquid.

[0016] The quantity of radioisotopes produced in a liquid target is very small (e.g., an isotope concentration in the target may be in the order of 10^{-12}), so it is important that the target body not introduce contaminants into the target material. Such contaminants would reduce the quantity of the available useful radioisotopes, and hinder the subsequent chemical processes in incorporating the radioisotope into the desired radiochemical.

[0017] Removal of the heat generated in the target is a significant problem that limits the magnitude of the incoming beam's current and hence, the production rate. Higher production rates are achieved if beams with higher currents can be used. Prior art target holders have been made of silver, which has a high thermal conductivity that allows heat to be quickly drawn from the liquid target. The silver target holders, however, often introduce impurities such as silver oxides that can react with or impede the reaction of the radiochemical formed in the target holder.

[0018] A description of water targets is provided in an article titled "Tantalum [^{18}O] Water Target for the Production of [^{18}F] Fluoride with High Reactivity for the Preparation of 2-Deoxy-2-[^{18}F]Fluoro-D-Glucose," by N. Satyamurthy, Bernard Amarasekera, C. William Alvord, Jorge R. Barrio, Michael E. Phelps, in Molecular Imaging and Biology, Vol. 4, No. 1, at 65–70 (2002). This article describes the use of tantalum for the body of the water target and discloses some of the disadvantages and problems of the prior art silver target assemblies. The article further discloses the lower heat conductivity of tantalum, along with its chemical inertness, radiochemical reactivity, and low induced activation. Figure 1 of the article illustrates that the target assembly is cooled by heat transfer into a cooling water plenum located inside the assembly. Test results using tantalum

show an average actual yield of 112.7 mCi/ μ A for the nine runs over 60 minutes in duration. This yield is 68.3% of the theoretical yield. None of the documented tests had a beam current above 40 μ A and the beam energy was at 10.8 MeV.

[0019] An example of target cooling is disclosed in United States Patent Number 5,917,874, titled "Accelerator Target," issued to Schlyer, et al. on June 29, 1999, which discloses a target **14** with radial cooling fins **28**. The Target **14** contains a sample **12** in the front side and a cooling system on the back side. The cooling system includes an integral solid cone **42** with a grouping of radial fins **28** disposed on the outer surface of the cone **42** to increase the surface area for cooling. A water jet **40a** is directed at the apex **42a** of the cone **42** from a single center inlet **40d**. The coolant **40a** flows along the cone **42** and radial fins **28**, through a plenum **40c**, and out a pair of outlets **40e**.

[0020] United States Patent Number 6,586,747, titled "Particle Accelerator Assembly With Liquid-Target Holder," issued to Erdman on July 1, 2003, discloses a target assembly **12** with two windows **62**, **64**. The target cavity **60** has a front window **62**, formed of Havar, through which the particle beam **17** passes. The target cavity **60** has a thin rear window **64**, formed of a thin section of the holder body **56**, formed of niobium, which separates the target cavity **60** from the cooling channel **74**. Transfer of the heat from the target cavity **60** is through the rear window **64** and by passing cooling fluid through the cooling block **68** and over the rear window **64**. The cooling block **68** is mounted to the holder body **56** and has support ribs **72** that form parallel cooling channels **74** through which the cooling fluid flows. The target cavity **60** is at an angle to the particle beam **17**, thereby allowing the particle beam **17** to pass through a greater thickness of the target fluid **54**, which allows for using higher energy particle beams **17**.

BRIEF SUMMARY OF THE INVENTION

[0021] According to one embodiment of the present invention, a target assembly is provided. The target assembly includes channels in which developed flow of a coolant removes the heat from the target liquid. In one embodiment, a pair of parallel channels provide cooling. In another embodiment, the target assembly is fabricated out of tantalum, which allows for higher current proton beams to be applied to the target liquid without reducing the life of the target assembly or introducing contaminants in the target liquid. In still another embodiment, the target chamber is shaped to promote natural circulation of the target liquid as it undergoes bombardment.

BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

[0022] The above-mentioned features of the invention will become more clearly understood from the following detailed description of the invention read together with the drawings in which:

Figure 1 is a front perspective view of a prior art target assembly;

Figure 2 is a rear perspective view of the prior art target assembly;

Figure 3 is a cross-sectional view of the prior art target assembly;

Figure 4 is a front perspective view of one embodiment of a target assembly;

Figure 5 is a cross-sectional view of one embodiment of a target assembly;

and

Figure 6 is a cross-sectional view of the upper cooling channel and the target chamber.

DETAILED DESCRIPTION OF THE INVENTION

[0023] An apparatus for containing and cooling a liquid target is disclosed. The apparatus, a target assembly **10**, has a chamber in which enriched water is irradiated with a proton stream.

5 **[0024]** Figures 4 and 5 illustrate one embodiment of the present invention. The target assembly **10** has a target body with a relatively solid outside surface with an upper flow channel **404** and a lower flow channel **406** through which cooling water can be provided. The target chamber **104'** has a front window **310** approximating a one-quarter circle, and the target chamber **104'** extends into the
10 target assembly **10** with a sloping, or canted, rear wall **512** to allow for expansion of a vapor jet adjacent to the beam strike area **312** of the entrance window **310**. The target liquid is introduced into the target assembly **10** through port **106**, located at the lower portion of the target chamber **104'** and extending into the front face **112** of the target assembly **10**.

15 **[0025]** In one embodiment, the target assembly **10** is fabricated of tantalum, which has superior oxidation resistance compared to silver, but poorer thermal conductivity. Silver has high thermal conductivity of 415 W/m-K, whereas tantalum has a lower thermal conductivity of 57 W/m-K. Target assemblies fabricated of silver encounter oxidation problems with beam currents above 60 μ A.
20 Target assemblies **10** of tantalum have been tested up to 100 μ A (1000 W at 10 MeV) and have provided excellent longevity and increased output at heretofore unattainably high production levels.

[0026] Figure 5 illustrates a section of the target **10** through one of two parallel channels **502**, **504**, **506**, **508**, each off center relative to the vertical
25 midplane of the target **10**. Each of the two channels are defined by 4 blind holes **502**, **504**, **506**, **508**, which, in one embodiment, are drilled into the target assembly **10**. In one embodiment, the 4 blind holes **502**, **504**, **506**, **508** are each 0.067" diameter and are approximately 0.180" off the vertical midplane of the target **10**.

[0027] In operation, the target liquid is introduced into the target chamber **104'** through the port **106**. Cooling water is pumped from the lower channel **406**, through the two parallel channels **502**, **504**, **506**, **508**, and into the upper channel **404**. The target liquid is irradiated and the heat is removed by the cooling water
5 flowing through the channels **502**, **504**, **506**, **508**. In particular, a high Reynolds number flow path through the two parallel channels **504**, **506** cool the horizontal upper condenser plate surface **514** and the canted back wall **512** inside the beam strike, thereby compensating for the low thermal conductivity of the tantalum target assembly **10**.

[0028] The target assembly **10** includes a target chamber **104'**, which is filled with enriched water via an inlet port **220** on the back side **212**. The target chamber **104'** is sealed with a window **310** adjacent the front face **112**. The inlet port **220** feeds an inlet channel **106**, through which the enriched water enters and fills the target chamber **104'**. The air pushed out of the target chamber **104'**
15 exhausts through the outlet port **222**. Before being irradiated, the enriched water completely fills the target chamber **104'**. The accelerator beam strikes the target chamber **104'** at a circular region **312** (the beam strike) in the lower portion of the chamber **104'**. The beam heats the window **310** and the enriched water in the immediate vicinity of the window **310**. The window **310** is typically Havar and is
20 elevated to a high temperature by the beam. The window **310** transfers some of its heat to the water, which is also being heated by the beam. The enriched water experiences localized boiling adjacent to the window **310** at the beam strike area **312**, which causes a jet of superheated steam to form. The jet moves upward, into a stable steam bubble in the top portion **514** of the target chamber **104'**. The
25 enriched water circulates in the target chamber **104'** from the target strike area **312**, to the top portion **514** of the target chamber **104'**, where it is condensed, down the back wall **512** and the side walls of the chamber **104'** and toward the front window **310**, where the enriched water re-enters the beam strike area **312** and is reheated, continuing the cycle.

[0029] The cooling water enters the lower channel **502** and passes through the channel **504** adjacent the rear wall **512** of the target chamber **104'**. The

cooling water, which is warmer after passing by the rear wall **512**, then passes through the channel **506** adjacent the upper wall **514** of the target chamber **104'** and then out of the target assembly **10** through the upper channel **508**. The cooling water progressively heats as it moves through the channels **502**, **504**, **506**, **508**, thereby presenting the enriched water at the back wall **512** with the coolest water possible. The differential temperature between the enriched water and the cooling water is maximized by having the cooling water enter at the bottom. Further, the developed flow of the cooling water allows for greater heat transfer from the target assembly **10**.

[0030] The embodiment of the target chamber **104'** illustrated in Figure 5 has a configuration that aids the cooling of the enriched water by allowing for natural circulation of the enriched water. In one embodiment, the function of containing the target liquid for irradiation is performed by the target chamber **104'** within the target body. In another embodiment, the function of containing the target liquid for irradiation is performed by the target chamber **104'**, which includes the arcuate upper wall **514** and the back wall **512**. In one embodiment, the function of cooling the target assembly **10** is performed by at least one cooling channel **506** adjacent to and parallel to the upper wall **514**, with the cooling channel **506** having developed flow. In another embodiment, the function of cooling the target assembly **10** is performed by at least one set of cooling channels **504**, **506** adjacent to and parallel to the back wall **512** and the upper wall **514**, respectively, with the cooling channels **504**, **506** having developed flow.

[0031] In one embodiment, the function of inducing fluid flow within the target chamber **104'** is accomplished by the shape of the target chamber **104'**. In another embodiment, the function of inducing fluid flow within the target chamber **104'** is accomplished with the front window **310** having a larger area than the beam strike area **312**, the curved upper wall **514**, and the canted back wall **512**.

[0032] In one embodiment, the flow is adjusted to 0.25 gpm through each of the two parallel channels **502**, **504**, **506**, **508** and for a 5 psi drop. The Reynolds number calculated for this configuration is 11799, indicating a truly turbulent

regime. The flow is fully developed in the slanted channel **504**, and nearly fully developed in the top horizontal channel **506**. The pressure available in the target assembly **10** is being used more efficiently than in the prior art. The pressure drop along the two channels **504**, **506** sums to 4.73 psi. These numbers also compare favorably with an inlet dynamic head of 0.04 psi, indicating that flow instabilities from entrance conditions are less likely. The target assembly **10** has heat transfer coefficients of 32,019 W/m²-K, owing to the turbulent diffusion of thermal energy. This gives much lower and more realistic temperature drops in the boundary layer, and a reasonable 3.81 degrees Celsius increase in water temperature over the course of the flow.

[0033] Figure 6 is a cross-sectional view illustrating one of the parallel upper channels **506** and the top surface **512** of the target chamber **104'**. The enriched water in the target chamber **104'**, in one embodiment, is pressurized to 600 psi. The circular cross-section of the channels **504**, **506** allows the channels **504**, **506** to be close to the surface of the target chamber to maximize heat transfer while still allowing the target chamber **104'** to contain an elevated pressure without rupturing. With the low heat transfer rate of tantalum, cooling efficiency is increased by locating the channels **504**, **506** as close as possible to the back and upper walls **512**, **514** of the target chamber **104'**.

[0034] The shorter conduction paths **504**, **506** and more optimal cooling enables operation of target assemblies **10** with materials such as tantalum, which are less desirable from the standpoint of thermal conductivity, but have superior chemical properties. The complexity of the target assembly **10** has also been reduced, compared to the prior art target assembly **110**.

[0035] Extensive testing of the illustrated embodiment of the target assembly **10** has been conducted. The tested target assembly **10** was constructed of tantalum. With 48 runs of over 60 minutes duration, the average actual yield of 130.7 mCi/ μ A. This yield is 84.5% of the theoretical yield, which is much greater than the yield achieved from the target assembly described in the Satyamurthy article.

[0036] The Satyamurthy article used an RDS-112 accelerator, which has a beam energy, after passing through all of the entrance foils, of approximately 10.8 MeV. At that energy, the theoretical yield of the ^{18}F production in ^{18}O enriched water is 165 mCi/ μA at saturation. In the bombardments over 60 minutes in duration (n=9), the average saturation yield obtained with the configuration of the target assembly disclosed in the Satyamurthy article was 112.7 mCi/ μA at saturation, or 68.3% of theoretical.

[0037] The tested target assembly **10** was operated with a gridded window support which intercepts beam current, so an additional correction factor of 0.91 was applied to the beam current. With this correction, the average saturation yield of the bombardments over 60 minutes in duration (n=48) was 130.7 mCi/ μA at saturation. The tested embodiment had currents of 60 to 100 μA . The accelerator these bombardments were performed with, the RDS Eclipse, has a beam energy of about 10.3 MeV after passing through all foils. At that lower energy than the accelerator used for the Satyamurthy experiments, the theoretical yield is 154.7 mCi/ μA at saturation. Therefore tested target assembly **10** achieves 84.5% of theoretical yield, even though the beam current is much higher than the target assembly used in the Satyamurthy article. This high yield with tantalum is an unexpected benefit. Although known in the art, the use of tantalum, in combination with the cooling system described herein, provides unexpected results considering the low heat coefficient of tantalum and the use of higher beam currents.

[0038] From the foregoing description, it will be recognized by those skilled in the art that a novel target assembly has been provided. The target assembly is fabricated of tantalum, which has superior oxidation resistance, and has cooling channels utilizing minimal conduction paths and high Reynolds number flows, which permits the target assembly to operate at high beam currents. The higher beam currents, along with the oxidation resistance, increases the performance and production capabilities over the prior art target assemblies.

[0039] While the present invention has been illustrated by description of several embodiments and while the illustrative embodiments have been described in considerable detail, it is not the intention of the applicant to restrict or in any way limit the scope of the appended claims to such detail. Additional advantages
5 and modifications will readily appear to those skilled in the art. The invention in its broader aspects is therefore not limited to the specific details, representative apparatus and methods, and illustrative examples shown and described. Accordingly, departures may be made from such details without departing from the spirit or scope of applicant's general inventive concept.